Reply to Reviewer 1’s Comments

This paper discusses the outcome of an improvement by the authors to the aerosol model (the Modal Aerosol Model 7 or MAM7) of the Community Earth System Model (CESM). The authors have successfully incorporated several sophisticated components to replace the more simplified counterparts in the current MAM7/CESM. These include a gas chemistry package, an aerosol thermodynamics module, and a new ionbased aerosol nucleation module. Besides, the authors have also performed a sensitivity simulation using altered emissions. The effort is to assess the performance of the CESM model after the introduction of these new aerosol and chemistry components.

I admire this effort that covers many detailed aspects of model prediction of aerosols and tropospheric chemistry. On the other hand, however, the paper is mostly about model development rather than science findings. Noticeably, all the model components introduced into the CESM have been developed previously and mostly used in other frameworks, thus what the paper describes is the outcome from coupling these components with a different model framework. This would still be invaluable. The work has provided an arguably better platform than the current CESM in modeling aerosol and chemistry interaction with climate. The evaluation of such a model improvement needs to be recorded in literature. A relevant question here is whether the authors would want to consider journals that specifically solicit works dealing with model techniques and evaluations such as Geoscientific Model Development. If not, I would suggest the authors reconsider the scope of the paper and focus more on science features in discussion. My (most general) comments here are for the authors to consider when making their revision, either for ACP or resubmitting to another journal.

Reply:

We thank the reviewer for positive comments. It is true that the model components such as ISORROPIA II and YU10 ion-mediated nucleation parameterization were developed previously and have been used in some other models. However, incorporating them into CESM/CAM5 is not a trivial effort, as it involves a substantial amount of recoding of some of those model components, developing new interface between them and the host model, and testing and evaluating against a large set of observational data. We agree with the reviewer that such work is invaluable and should be recorded in literature.

We also thank the reviewer for a suggestion to submit our paper to Geoscientific Model Development (GMD). However, prior to our submission to ACPD, we have considered several possible journals including GMD and believe that ACP is the best journal for our paper, as our work fits well into the scope of work published by ACP.

The authors used the fully coupled CESM configuration to run their simulations. This is a configuration includes a full ocean GCM coupled with the atmospheric model along with other components in a transient mode. Surprisingly, with this configuration, all the simulations were integrated for only one year. In the discussion of modeled results, the authors spent quite an effort on changes of many climate variables from surface air temperature to wind speed. Arguably, this is not adequate because of the short integration time and very long ocean response.

Note that the model was cold started and forced by an introduced new and different forcings than the standard 1850-2000 run (it is understood that the last output of this run was used as initial condition for the simulations). In my opinion, the emphasis of the effort should be, as stated by the authors, to assess the aerosol and chemistry predictions introduced by the new model components. Such an effort, however, would likely suffer from the first year response of modeled transient climate after a cold start, which involves interactions between aerosol, air chemistry and meteorology/climate as well. It would be much better if the authors use a configuration using prescribed SST, run simulations for at least three years, and then remove all the irrelevant discussions of climate variables (the changes are hardly to identify anyway from Table 3) from the paper but those of chemistry and aerosol features.

Reply:

While the initial conditions for ice model and ocean model are from CESM default settings, the initial conditions for land model are the output from NCAR CESM B_1850-2000_CN run. The initial conditions for CAM5 are derived from a 10-yr (1990-2000) CAM5 standalone simulation with the MOZART chemistry provided by NCAR. A 1-year (January 1-December 31, 2000) CESM/CAM5 simulation using NCAR's CESM B_1850-2000_CAM5_CN component set is performed as spinup to provide the initial conditions for meteorological variables and chemical species that are treated in both MOZART and CB05_GE. An additional 3-month (October 1-December 31, 2000) CESM/CAM5 simulation based on a 10-month (January-October, 2000) CESM/CAM5 output using initial conditions from NCAR's CESM B_1850-2000_CAM5_CN is performed as spinup to provide initial conditions for chemical species that are treated in CB05_GE but not in MOZART. So, the model was not cold started for atmospheric and land models. We have clarified this in the revised paper.

We agree with the reviewer that the 1-yr simulation time is a bit too short for global model simulations and the results may be more robust with longer simulations and prescribed SST. However, we believe that the changes in most radiation, aerosol, and cloud-related variables are caused mainly by changes in the model representations. We have performed the student's t-test, which verified this. Therefore, we kept Table 3, but we provided info from our t-test in Table A1 in the supplementary material.

To address the reviewer's comment, we have performed additional simulations for 2001-2005 with prescribed SST and analyzed the results in the revised paper. To compare predictions with prescribed SST and in a fully-coupled mode, we also performed a 5-yr simulation for 2001-2005 using fully-coupled CESM. These new results are shown in Tables 6-7, Figures 9-10, and a new discussion section (i.e., Section 6).

The improvements in terms of aerosol and chemistry model predictions are mostly limited in comparison to observations (see Table 4), while the conclusion drawn in abstract and Conclusion section about this is much too optimistic to me. Realizing the very high demanding in computation of this new model, the best purpose this effort could serve is to provide certain information for the improvement of simplified schemes, because practically it would come to the usage of the latter types to conduct aerosol chemistry-climate interaction simulations. Therefore, the emphasis perhaps should be on the relative differences between various model configurations.

Reply:

We agree that it is interesting to compare the relative differences between various model configurations, our submitted paper actually includes extensive discussions on such differences (e.g., most of the original Section 5 describes such differences along with all original Figures 1, and 3-9). The evaluation with observations would attest if the improved model representations improve the model performance, and the performance statics shown in the original Tables 3 and 4 also illustrated the differences in model predictions using different model configurations. Such differences continued to be the main focus of our discussions in the revised paper, as shown in the revised Section 5 and newly added Section 6, Figures 1, and 3-10, and Tables 3, 4, 6, and 7.

Given rapid advancement of computer powers from Teraflops to Petaflops, we believe that in near future, long-term aerosol chemistry-climate interaction simulations can be performed using CESM with more comprehensive aerosol and chemistry treatments. Therefore, the evaluation presented in this work would provide a benchmark to assess the capability of the improved model vs the original model with the simplified schemes and/or to provide rational for any further improvement in the future.

There is an argument that the emission uncertainty could be the reason for much of the remaining problem of the new model. This is also too simplistic to me. By introducing those sophisticated model components, the uncertainty is multi-dimensional (a good example is the scale inconsistency of fast chemistry in the current model). In addition, the sensitivity simulation using alternative emissions were not clearly explained in the paper.

Reply:

We agree with the reviewer that the uncertainty is multi-dimensional when introducing these sophisticated model components. We did not state or imply in the manuscript that "the emission uncertainty could be the reason for much of the remaining problem of the new model." What we mentioned was "some large biases are caused by inaccuracies in the emissions of CO, SO_2, BC, OC, and NH_3,". As a matter of fact, we indeed indicated that "Additional uncertainties exist in the model treatments" in the conclusion section (last paragraph) in our submitted paper, in addition to the uncertainty in the anthropogenic emissions. We provided several examples in that section, e.g., uncertainty in the dust emission treatment, nucleation parameterizations, SOA module, cloud microphysics schemes and aerosol-cloud interaction parameterizations. The uncertainty may also stem from other model settings such as the use of a coarse grid resolution and a large time step for solving chemical ODEs. Emission uncertainty could be one of the reasons for model bias, which can be estimated through the sensitivity simulation using alternative emission inputs.

To address the reviewer's comment, we have added a few more factors that may contribute to model uncertainties such as the use of a coarse grid resolution and a large model time step for solving the chemical system in the last paragraph of the conclusion section.

The current discussions appear to have too many leads, while detailed connections among various chemical or physical processes were not clearly analyzed. For example, simulations of tropospheric chemical features seem being improved much significantly over Europe than other places, the reasons behind this, however, has not been explained or stated clearly. More quantitative comparisons between precursors and products in different continents (could be selective) would help (note that the information in Table 4 is incomplete, see specific comment).

Reply:

Given the comprehensive evaluation that we performed, it is not possible to do quantitative comparisons between precursors and products in different continents. However, we indeed selected representative continents for such discussions. For example, in Section 5.5 we discussed the main reason for the large NMB of O_3 predictions over Europe in MAM_NEW_A and showed additional seasonal performance statistics over Europe in Table 5.

The chemical species included in the observational networks in East Asia are not as many as in Europe and CONUS. We have included major chemical species whose observations are available from East Asia in Tables 4 and 6.

To address the reviewer's comment, we included additional evaluations for NO_2 and O_3 over East Asia using limited observations in those tables.

Several Specific Comments.

1. How many variables are prognostic and included in tracer advection? This would be useful for the reader to estimate the workload of the new model.

Reply:

There are a total of 139 prognostic species included in tracer advection. This information has been added in the revised paper.

2. How were the photolysis rates calculated, using any J-table (note, not the nucleation rate)?

Reply:

The photolytic rates are calculated based on Lamarque et al. (2012) which used a combination of a lookup table and online calculation. Photolytic rates for wavelengths larger than 200 nm are calculated using a lookup table, which is based on the Stratosphere, Troposphere, Ultraviolet (STUV) radiative transfer model. At wavelengths less than 200

nm, the wavelength-dependent cross section and quantum yields for each species are specified and the transmission function is calculated explicitly for each wavelength interval.

The Lamarque et al. (2012) reference has been added in the revised paper.

3. P. 27720, L17-19, ": : :mass accommodation coefficient: : :can be measured: : :", the authors might want to indicate the high uncertainty in this type of measurements, as being demonstrated later in the paper by a difference in orders of magnitude in the adopted values of sulfuric acid and others.

Reply:

The reviewer is correct that such measurements are highly uncertain. This uncertainty has been indicated in the revised paper.

4. P.27728, it may help if the authors could further explain the derivation of the initial condition for the model, was the ending result of B1850-2000 run used?

Reply:

Please see our reply to the general comment on this.

5. Section 3.3, although the evaluation procedure might have been explained in detail in the cited publication, it would still be useful if the authors could explain briefly about this procedure, for instance, whether the data of 2001 or multi-year average data were used in the comparison, etc..

Reply:

All observational data used for evaluating 2001 simulations are based on 2001 only except for particle formation rates that are based on different years during the period of 1998-2002 compiled from Kulmala et al. (2004) and Yu et al. (2008). All observational data used for evaluating 2001-2005 simulations are based on 2001-2005.

This information has been added in the revised paper.

6. P.27732, Section 4, how did the modeled aerosol results in the MAM_SIM differ from those in Liu et al. (2012)?

Reply:

The results from MAM_SIM in Section 4 are not comparable to those in Liu et al. (2012) for several reasons. First the datasets used to generate initial conditions are different in our work and Liu et al. (2012). We use the B_1850-2000_CAM5_CN configuration for

MAM_SIM, whereas Liu et al. (2012) used CAM standalone configuration. Second, our MAM_SIM was performed only for 1-yr period, i.e., 2001 whereas the simulation results in Liu et al. (2012) were based on 5-year average. Finally, the emissions used in our simulations are different from those in Liu et al. (2012). Those differences in model configurations and inputs would result in different aerosol predictions.

To address the reviewer's comment, we compared the global burden of major gaseous and aerosol species from MAM_SIM_5Y with those from Liu et al. (2012) since similar configurations and time period are used for both simulations, although they used different emission inventories. This comparison has been added in the revised paper in Section 6 and Table 8.

7. Many of the gas chemistry comparisons were done by comparing the new model results with those of MAM_SIM, this is not informative in my opinion. Many of the chemical fields in MAM7 are prescribed using climatological data derived from MOZART model or alike based on my understanding, and this is done for reason. The authors should compare the chemistry features to the "mother" model of chemical fields used in MAM7, such information would be useful for the aerosol modelers to decide whether they should use alternative climatology, and if not, which aspect in the current climatology need to be improved.

Reply:

We believe that comparisons of results from the improved model with those of MAM_SIM are useful, as they show differences in model predictions due to different gas-phase chemical mechanisms.

We agree that comparison of results from the improved model with those of the "mother" model of chemical fields used in MAM7 would also be useful for the reason stated by reviewer. We have therefore compared prescribed OH, HO_2, NO_3, and O_3 in MAM_SIM with their prognostic predictions from the improved model and added a new Figure (Figure 1b) for this comparison in the revised paper.

8. P.27733, the last paragraph and other places, is SO4= here in aerosol or aqueous phase?

Reply:

SO_4^{2-} is in the particulate phase. This has been indicated clearly when we define it for the first time in Section 1 in the following statement:

Inorganic aerosols comprise 25-50% of fine aerosol mass (Heintzenberg, 1989), which mainly includes sulfate (SO_4^{2-}), ammonium (NH_4^+), nitrate (NO_3^-), chloride (Cl^-), and sodium (Na^+).

9. P.27735, L20, "The aforementioned changes: : :", changes due to what?

Reply:

The changes are due to new gas-phase chemistry implemented in the model and the feedbacks to meteorology and radiation through the climate system. This has been indicated in the revised paper.

10. P.27746, L25, "CCN: : : cmˆ(-2)", is this referred to the column loading?

Reply:

Yes, this is referred to the column loading. To clarify this, we have changed "CCN at a supersaturation of 0.5% by…" to "Column CCN at a supersaturation of 0.5% by…"

11. P.27747, last two lines and following, "Large biases : : :due to uncertainties in model input (e.g., meteorology and emissions): : :", this deserves a thorough analysis, or at least citations to support this statement.

Reply:

To address the reviewer's comment, we have added more discussions on the emission impacts in Section 5.6 in the revised paper.

12. P.27749, 5.6; it is not quite clear that what emission inventory the authors had used in this simulation, and what the differences are between the alternative emission inventory and the default one. If the authors selectively adjusted emissions for certain species, what are these species then and how was the adjustment done and based on which works?

Reply:

As indicated in lines 23-29, P 27728 and lines 1-2, P 27729 of our ACPD paper, the default emission inventory used in all simulations except for MAM_NEW/EMIS is the 2001 emission file of Zhang et al. (2012). Table 2 of Zhang et al. (2012) showed the sources of this 2001 detailed emission inventory. Those adjusted emissions used in MAM_NEW/EMIS are adjusted based on the comparison with the emission inventories from the Representative Concentration Pathways (RCPs), the MOZART version 4 (MOZART-4), the Reanalysis of the TROpospheric chemical composition (RETRO), the Global Fire Emissions Database (GFED) version 2, and preliminary evaluation of CESM/CAM5.1 with modified and new gas and aerosol treatments using available observations.

To address the reviewer's comment, we have referred the readers to Table 2 of Zhang et al. (2012) for details in this 2001 emission inventory (This Table 2 is provided below for the reviewer's convenience).

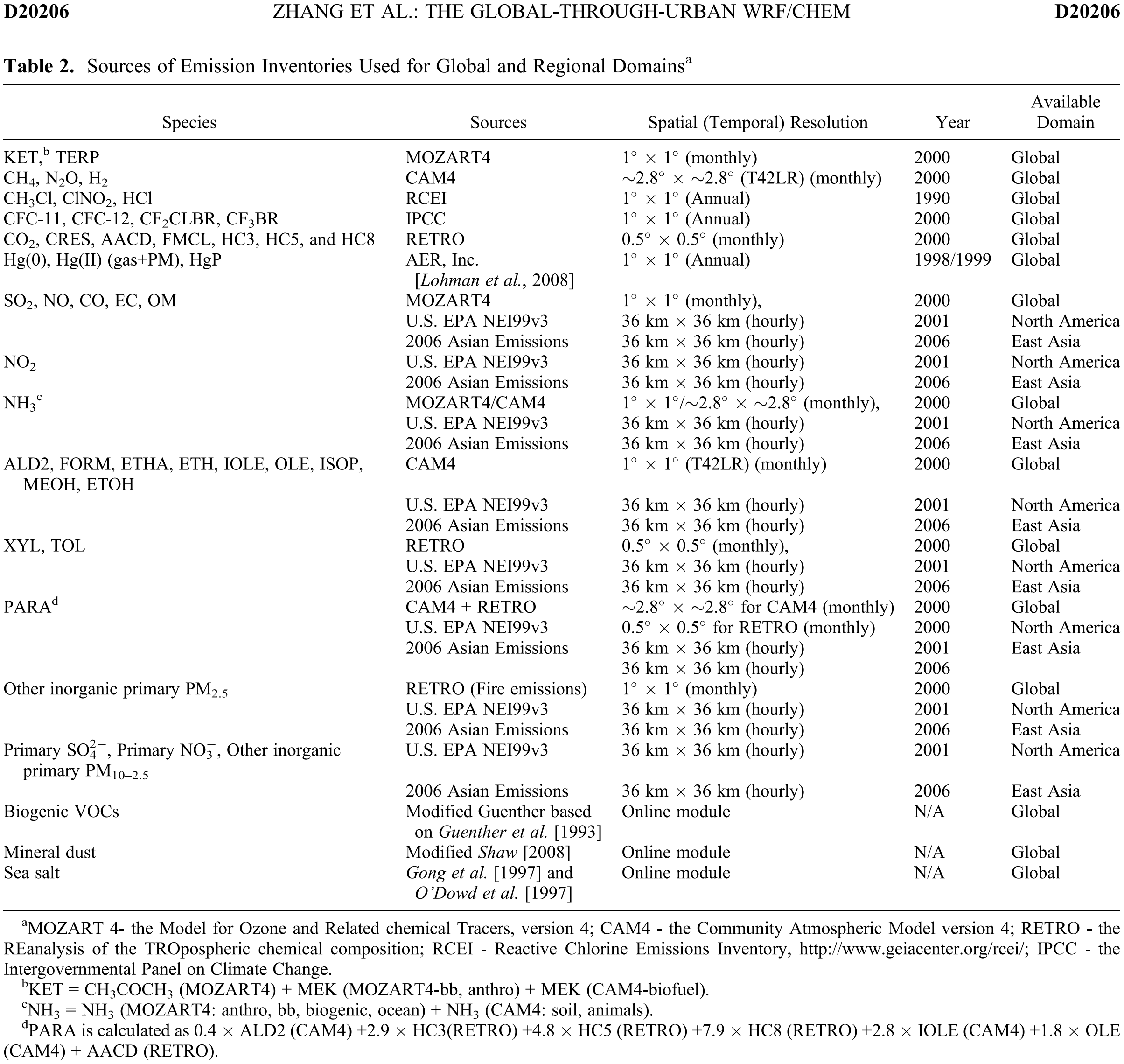

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Table 2. Sources of Emission Inventories Used for Global and Regional Domains[a]

Species	Sources	Spatial (Temporal) Resolution	Year	Available Domain
KET,[b] TERP	MOZART4	1° × 1° (monthly)	2000	Global
CH_4, N_2O, H_2	CAM4	~2.8° × ~2.8° (T42LR) (monthly)	2000	Global
CH_3Cl, $ClNO_2$, HCl	RCEI	1° × 1° (Annual)	1990	Global
CFC-11, CFC-12, CF_2CLBR, CF_3BR	IPCC	1° × 1° (Annual)	2000	Global
CO_2, CRES, AACD, FMCL, HC3, HC5, and HC8	RETRO	0.5° × 0.5° (monthly)	2000	Global
Hg(0), Hg(II) (gas+PM), HgP	AER, Inc. [*Lohman et al.*, 2008]	1° × 1° (Annual)	1998/1999	Global
SO_2, NO, CO, EC, OM	MOZART4	1° × 1° (monthly),	2000	Global
	U.S. EPA NEI99v3	36 km × 36 km (hourly)	2001	North America
	2006 Asian Emissions	36 km × 36 km (hourly)	2006	East Asia
NO_2	U.S. EPA NEI99v3	36 km × 36 km (hourly)	2001	North America
	2006 Asian Emissions	36 km × 36 km (hourly)	2006	East Asia
NH_3[c]	MOZART4/CAM4	1° × 1°/~2.8° × ~2.8° (monthly),	2000	Global
	U.S. EPA NEI99v3	36 km × 36 km (hourly)	2001	North America
	2006 Asian Emissions	36 km × 36 km (hourly)	2006	East Asia
ALD2, FORM, ETHA, ETH, IOLE, OLE, ISOP, MEOH, ETOH	CAM4	1° × 1° (T42LR) (monthly)	2000	Global
	U.S. EPA NEI99v3	36 km × 36 km (hourly)	2001	North America
	2006 Asian Emissions	36 km × 36 km (hourly)	2006	East Asia
XYL, TOL	RETRO	0.5° × 0.5° (monthly),	2000	Global
	U.S. EPA NEI99v3	36 km × 36 km (hourly)	2001	North America
	2006 Asian Emissions	36 km × 36 km (hourly)	2006	East Asia
PARA[d]	CAM4 + RETRO	~2.8° × ~2.8° for CAM4 (monthly)	2000	Global
	U.S. EPA NEI99v3	0.5° × 0.5° for RETRO (monthly)	2000	North America
	2006 Asian Emissions	36 km × 36 km (hourly)	2001	East Asia
		36 km × 36 km (hourly)	2006	
Other inorganic primary $PM_{2.5}$	RETRO (Fire emissions)	1° × 1° (monthly)	2000	Global
	U.S. EPA NEI99v3	36 km × 36 km (hourly)	2001	North America
	2006 Asian Emissions	36 km × 36 km (hourly)	2006	East Asia
Primary SO_4^{2-}, Primary NO_3^-, Other inorganic primary $PM_{10-2.5}$	U.S. EPA NEI99v3	36 km × 36 km (hourly)	2001	North America
	2006 Asian Emissions	36 km × 36 km (hourly)	2006	East Asia
Biogenic VOCs	Modified Guenther based on *Guenther et al.* [1993]	Online module	N/A	Global
Mineral dust	Modified *Shaw* [2008]	Online module	N/A	Global
Sea salt	*Gong et al.* [1997] and *O'Dowd et al.* [1997]	Online module	N/A	Global

[a] MOZART 4- the Model for Ozone and Related chemical Tracers, version 4; CAM4 - the Community Atmospheric Model version 4; RETRO - the REanalysis of the TROpospheric chemical composition; RCEI - Reactive Chlorine Emissions Inventory, http://www.geiacenter.org/rcei/; IPCC - the Intergovernmental Panel on Climate Change.

[b] KET = CH_3COCH_3 (MOZART4) + MEK (MOZART4-bb, anthro) + MEK (CAM4-biofuel).

[c] NH_3 = NH_3 (MOZART4: anthro, bb, biogenic, ocean) + NH_3 (CAM4: soil, animals).

[d] PARA is calculated as 0.4 × ALD2 (CAM4) +2.9 × HC3(RETRO) +4.8 × HC5 (RETRO) +7.9 × HC8 (RETRO) +2.8 × IOLE (CAM4) +1.8 × OLE (CAM4) + AACD (RETRO).

13.Table 4. Noticeably, the listed regions for each variable are not always the same, for example, East Asia numbers were only listed for CO and SO2, especially not for ozone and many other gaseous and aerosol species. The authors might want to list the results from a same collection of regions.

Reply:

The chemical observations over East Asia are very limited, and they only include surface concentrations of CO, SO_2, NO_2, O_3, and PM_{10}.

To avoid confusion, we have indicated this in the revised paper.

Figures. The color scale in several figures was not always selected to show the necessary details, e.g., Fig. 7 and 8.

Reply:

We have modified the color scale used in Figures 7 and 9 in the revised paper. Note that the original Figure 8 from 1-yr (2001) simulation was replaced by Figure 9 from the 5-yr (2001-2005) simulation, in which appropriate scales were used for all plots.

Reply to Reviewer 2's Comments

This paper discusses a collection of improvements relevant to the representation of aerosols in CESM/CAM5. The present paper represents a large collection of efforts and as such is an important contribution to the model development. However, the use of CESM in a fully coupled climate mode, while allowing for full climate feedbacks, also makes the comparison of very short simulations meaningless. The main reason is that two simulations that are slightly different (whether form initial conditions or slightly different forcings/emissions/chemistry) will generate meteorologies that are significantly different, but this only represents the inherent noise of the climate system. In the configuration used in this study, simulations of at least 20 years (and probably quite longer since some of those changes are relatively minor) would be necessary to start seeing differences that are above the natural variability of the system. As written, the paper cannot be considered for publication. I therefore suggest that the authors focus on the chemistry aspect of the study, and perform short simulations in which the meteorology is not affected by the changes in chemistry, or simulations with fixed SSTs (which would probably need to be on the order of 5-10 years). The length of the simulation should be defined such that the response in the system is above the natural variability of the reference case. Since I find that the paper will have to go over major revisions, I have only included a few additional comments.

Reply:

We thank the reviewer for constructive comments. While we agree with the reviewer that results from longer time simulations would be more robust, we believe that the 1-year simulations provide useful information for model development and sensitivity study. The differences among those 1-yr simulations are indeed mainly caused by changes in model treatments, rather than the inherent noise of the climate system. This was verified by performing t-test for simulation pairs with different model configurations.

To address the reviewer's comments, we performed 5-yr simulations with two different gas-phase chemical mechanisms (SIM and CB05_GE) both with prescribed SST. Those results are added in a new section (i.e., Section 6), Tables 6-7 and Figures 9-10.
In addition, we performed a 5-yr simulation with the CB05_GE gas-phase chemistry using fully-coupled CESM/CAM5 and compare its performance with the same configuration but with prescribed SST. The results are shown in Tables 6-7 and discussed in Section 6.

Other comments

1. The paper would benefit from the addition of simple diagnostics such as global budgets and lifetimes

Reply:

Since the removal rates of some species were not included in the CESM/Cam5 output, it is not possible to accurately estimate the global budgets and lifetimes. To address the reviewer's comments, we have calculated global burdens of major chemical species, and compared them with those of previous studies in Section 6 and Table 6 in the revised paper.

2. Section 2.2.2: it seems that a major limitation in the evaluation of various methods is the unavailability of observations. Please comment.

Reply:

Despite limited data, observational data of new particle formation rates (J) are indeed available for evaluation of model predictions. As described in Section 3.2, we used the collected observations from Kulmala et al. (2004) and Yu et al. (2008) for model evaluation. Such data exist during different years over different regions. The evaluation results are shown in Table 4.

3. Section 2.2.4, line 23: where does the HCl come from?

Reply:

Simulated HCl mixing ratios result from HCl emissions and gas-particle partitioning of total chloride. This has been indicated in the revised paper.

4. Page 27728, line 24: Please add table with emissions

Reply:

Table 2 in Zhang et al. (2012) provided such detailed information regarding emissions used in this work. We believe that the authors are discouraged by any journals to repeat the same information that has been published. To address the reviewer's comment, we have referred the readers to this table in Zhang et al. (2012) in the revised paper.

5. Section 4: tables 3 and 4 contain too much information. It would really help to provide this information in an easier format, maybe through bar graphs.

Reply:

While bar graphs can generally show well model performance, many bar plots are needed for the evaluation that we performed over different regions for many meteorological variables and chemical species. For some variables, the differences in performance statistics are small, which may not be clearly displayed in bar graphs. Considering the above reasons, we believe that the table summary of the performance statistics over different regions for many variables is the most appropriate and concise format to present our results.

6. Page 27731, line 5: what is the role of optical properties and water uptake on AOD biases?

Reply:

The aerosol optical properties are defined for each mode of the MAM based on Ghan and Zaveri (2007). Uncertainty in hygroscopicity of aerosol components and treatments of water uptake can result in uncertainty in AOD prediction.

To address the reviewer's comment, we have added above explanation in the revised paper.

7. Page 27735, lines 29-30: what are the % with respect to?

Reply:

The percentage differences are with respect to MAM_SIM. This was indeed indicated in line 25, "compared with MAM_SIM,…" in our ACPD paper.

Reply to Reviewer 3's Comments

The authors have implemented the gas-phase photochemical mechanism (CB05-GE), an ion-mediated nucleation parameterization, and an inorganic aerosol equilibrium module (ISORROPIA) to the existing framework of CESM/CAM5.1-MAM7 model. The performance of the updated CAM5.1 model is evaluated for the full year of 2001. Since one of the objectives of this work is to improve the global predictions of inorganic aerosols, it is critical that this is done correctly. Unfortunately, there are several significant technical and scientific issues regarding the implementation of the inorganic aerosol equilibrium module as outlined below in specific comments. These issues must be clearly addressed before the present work can be considered an improvement to the existing CAM5.1 model. In my opinion this will require major changes to the existing implementation of the inorganic aerosol partitioning calculations as well as model evaluation. I therefore recommend that the manuscript be rejected in the present form.

Reply:

We thank the reviewer for constructive comments. However, it appears to us that the reviewer may have misunderstood our implementation of ISORROPIA II into CAM5 and several related issues. We respectfully disagree with the suggested major changes to the existing implementation and the rejection of our paper suggested by the reviewer, as they are based on misunderstanding or somewhat incomplete understanding of our work. Below we clarify those issues.

Specific Comments

1. The authors have implemented thermodynamic equilibrium for the fine aerosol modes (sub-micron) only, while the coarse sea-salt and dust aerosol modes have been completely ignored. The reason given for this is oversimplification is that the coarse modes are typically not at equilibrium and that dynamically solving the coarse mode non-equilibrium system (together with fine modes) is computationally expensive. There have been a number of studies, including Hu et al. (2008) (on which the corresponding author Y. Zhang was a co-author) that have shown that equilibrium approach (and even the hybrid method) fails to predict the distribution of semi-volatile species (NH4, NO3, and Cl) because of the equilibrium and internal mixture assumptions. Thus, simply ignoring the problem because it is difficult to solve is not an acceptable solution, especially since there are a few computationally efficient methods in the literature that can overcome this difficulty with varying degrees of efficiency and accuracy – for example see Jacobson (2005), Zhang and Wexler (2006), and Zaveri et al. (2008). Therefore, the present implementation of ISORROPIA in CAM5.1 cannot really be considered as an improvement. To the contrary, it is erroneous and will lead to meaningless results even though they may fortuitously appear to compare well with observations.

Reply:

We respectfully disagree with the reviewer's statements regarding our implementation of ISORROPIA II.

As indicated by one of the reviewers (i.e., Dr. Shaocai Yu of the US EPA), "The incorporation of ISORROPIA II into CAM5.1 significantly advances aerosol treatments in current global and Earth system models." Note that CAM5.1 only treats aerosol thermodynamics that involves sulfate and ammonium and does not treat those for nitrate and cloride. Considering aerosol thermodynamic treatments for all volatile species using ISORROPIA represents a significant improvement. To our best knowledge, our model is the only global climate/Earth system model that includes ISORROPIA II, although several other global models include ISORROPIA, which is an older version of ISORROPIA II. The main differences between the two versions lie in that ISORROPIA II accounts for the impact of crustal species on aerosol thermodynamics, which is not accounted for in ISORROPIA.

Hu et al. (2008) indeed showed that bulk equilibrium approaches fail to predict the distribution of semivolatile species (e.g., NH_4^+, Cl^-, andNO_3^-) due to equilibrium and internal mixture assumptions. However, this was only true for two test cases (Hong Kong and Tampa Bay) shown in their paper, both cases occurred in the coastal areas where seasalt and nitrate concentrations in the coarse mode are relatively high, which may not represent the vast areas of the whole global domain. In CAM5, by default, it does not use the equilibrium approach for gas-to-particle mass transfer. Instead, it uses a kinetic approach to simulate condensation of several species on the surface of fine and coarse particles and inorganic aerosol thermodynamics of sulfate and ammonium for fine-mode particles.

As also indicated by Dr. Yu, nearly all regional and global models only consider thermodynamics for fine particles and neglect that for coarse particles. Not until recently, kinetic approach was considered for coarse particles in a regional model, i.e., CMAQ 4.7 and newer versions. This is supported by the well-accepted assumption that gas-to-particle mass transfer can reach thermodynamic equilibrium with a short time scale under most ambient conditions. Further, more than 90% of current regional/global models treat aerosols as internal mixtures. While the kinetic approach for both fine and coarse-mode particles is desirable, it is computationally much more expensive than equilibrium approach (by factors of 2-4 based on Hu et al. (2008)), thus the kinetic approach is appropriate for short-term model applications over regions with significant coarse particles when computational resources are permitted. Earth system models, on the other hands, are designed to run for multiple decades and a tradeoff must be considered between accuracy and computational efficiency, the latter is critical for application of such models on an order of 30-50 years or longer. While a few computationally-efficient methods for kinetic gas-to-particle mass transfer do exist, none of those have been applied for multi-decadal climate/Earth system applications. Their appropriateness for multi-decadal applications needs to be examined before they can be considered for implementation into global/Earth system models for such long-term model applications.

2. It is well known that, under stable conditions, inorganic aerosols can exist as completely solid (at low relative humidity), mixed solid and liquid phases (at moderate RH) and completely liquid (at high RH), depending on the mutual deliquescence relative humidity of a given multicomponent aerosol. They can also exist in completely liquid metastable state depending on

the aerosol processing history and hysteresis effects. However, it is not clear how the phase state of the inorganic aerosols is treated in the present implementation of ISORROPIA. The only place I found a mention of this process in the entire manuscript is in Table 1, where it is simply stated that ISORROPIA aerosol thermodynamics was evaluated under metastable conditions. It therefore appears that phase transitions and hysteresis effects were not even considered in the present study. This is again a significant shortcoming in a global simulation where aerosols may encounter the full spectrum of relative humidity and processing histories depending on the geographyical location and meteorological conditions. Simply assuming metastable conditions under all relative humidity conditions at all the time is a gross oversimplification and cannot be considered an improvement (as several other global models have done this or are already doing something similar). The authors should therefore implement a treatment for hysteresis to allow proper investigation of the effects of phase transitions on inorganic aerosol partitioning.

Reply:

For applications of nearly all regional and global models such as CMAQ and GEOS-Chem, the metastable condition is a commonly-accepted assumption. This is supported by the fact that most ambient conditions are in metastable conditions and the metastable condition assumption represents the best compromise between accuracy and computational efficiency, the latter is an important factor for large scale, long-term applications. ISORROPIA II does offer an option (i.e., stable conditions) to simulate phase transitions and hysteresis effects, which may be important under some conditions (e.g., RH < 50% for nitrate, Fountoukis et al., 2009). However, based on the global annual mean RH values for the 2001 simulation, most regions have RH values > 60-70% (exceptions are over desert/arid regions such as Australia, the northern Africa, Arabian Desert, northwestern China, and western U.S.). We chose metastable conditions for the aforementioned reasons, which are particularly true for multi-decadal applications of CESM/CAM5.

To address the reviewer's comment, we have performed an additional 2001 simulation with ISORROPIA II under stable conditions and compare the results with those under metastable conditions, see Table 4 and Figure 8 in the revised paper. These results showed that on a global scale, the differences in results between stable and meta-stable conditions are overall insignificant (4.2%, 12.8%, -0.013%, and 2.0% for SO_4^{2-}, NH_4^+, NO_3^-, and Cl^-, respectively). Such a comparison further justifies our choice of metastable conditions for global scale long-term applications.

3. The treatment of phase transition calculations in ISORROPIA is also somewhat problematic. It is my understanding that ISORROPIA does not rigorously solve solid-liquid equilibria, but rather tries to approximate it using an ad-hoc approach. For instance, Ansari and Pandis (1999) clearly showed that ISORROPIA has difficulty in reproducing the complex multistage deliquescence behavior and the associated water content in stable multicomponent aerosols due to many of the simplifying assumptions. More recently, Zaveri et al (2008) also showed that ISORROPIA predictions (stable solutions) had large errors compared to the benchmark thermodynamic model AIM (Wexler and Clegg, 2002) under low and moderate relative humidity conditions. Thus, even if the authors implement hysteresis in CAM5.1, ISORROPIA may still

not be able to correctly capture the phase transitions themselves and hence their effects on dynamic gas-particle partitioning to size distributed aerosols.

Reply:

ISORROPIA II has been implemented in GEOS-chem, and is proven to be capable of predicting observed gas-aerosol partitioning (Fountoukis and Nenes, 2007). Our evaluation results using observations also showed that ISORROPIA II is capable of simulating gas-particle partitioning in the Earth system model.

To address the reviewer's comment, we have run ISORROPIA version 1.7 (which is very similar to the version used in Zaveri et al., 2008) and ISORROPIA II box model under four conditions tested in Zaveri et al. (2008), i.e., Cases 1, 2, 8, and 9. The RH value used in Cases 1 and 2 is 30%, and those used in Cases 8 and 9 are 55% and 52%, respectively. Those results are compared with AIM results from Zaveri et al. (2008) in Table 1 in the supplementary material to this reply.

We compared the above model results with AIM and ISORROPIA shown in Figures 1 and 3 in Zaveri et al. (2008). As shown in Figure 1, for Case 1, the predictions from ISORROPIA II and ISORROPIA are very close to those from AIM for all species except for NH_4NO_3(s), for which ISORROPIA II and ISORROPIA give a lower value than AIM (~28.8 nmol m^{-3} vs. 50 nmol m^{-3}). For Case 2, gaseous species such as NH_3, HNO_3, and HCl predictions from ISORROPIA II are closer to those from AIM compared with ISORROPIA, although HNO_3 predicted by ISORRPIA II is about 37% higher than AIM. While predictions of $NaNO_3$(s) and Na_2SO_4(s) from ISORROPIA and ISORROPIA II agree well with those from AIM, that of NH_4NO_3(s) is ~21% lower than that of AIM. Compared to AIM, ISORROPIA II gives closer agreement to AIM except for Na_2SO_4(s). For Case 8, ISORROPIA II gives much closer agreement to AIM HNO_3 prediction (140 nmol m^{-3}) than ISORRPIA (163.7 vs. 236.0 nmol m^{-3}, respectively). Compared with ISORROPIA, ISORROPIA II also gives closer agreement with AIM predictions for NH_3, H_2O, NH_4^+, NO_3^-, Cl^-, and NH_4Cl(s). For Case 9, compared to ISORROPIA, ISORROPIA II gives closer agreement to AIM prediction for all species expect for $(NH_4)_2SO_4$(s), NH_4NO_3(s), and Na_2SO_4(s). These comparisons indicate that ISORROPIA used in Zaveri et al. (2008) may contain some bugs/inaccuracies. The latest version, i.e., ISORROPIA II, is more capable of predicting aerosol phase transition than ISORROPIA.

4. In section 2.2.4, line 25, the authors list a set of values for mass accommodation coefficients for H2SO4, NH3, HNO3, and HCl. This is very puzzling, because the mass accommodation coefficients are only needed if the gas-particle partitioning is done dynamically (kinetically) as opposed to the equilibrium assumption used in the present work. If the equilibrium assumption is enforced then the effects of differences in mass accommodation coefficients on the distribution of semivolatile species among different size particles will vanish by definition. Trying to somehow capture the kinetic effects with a purely equilibrium model violates the basic concept of thermodynamic equilibrium for semi-volatile species.

Reply:

The reviewer may have misunderstood the model treatment for gas-to-particle mass transfer in CAM5. It is not based on the equilibrium approach. Instead, it is based on a kinetic approach using the gas-to-particle mass transfer expressions of Seinfeld and Pandis (1998), although the non-equilibrium process is not treated for coarse particles. The condensation of species on the surface of fine and coarse mode particles is simulated by using a set of values for species-dependent mass accommodation coefficients for H_2SO_4, NH_3, HNO_3, and HCl, although such a condensation is assumed to be an irreversible process by default in CMA5.

This has been clarified in the revised paper.

5. The values of the mass accommodation coefficients used are also very surprising. Some of them appear to be way too low (they are 0.02, 0.097, 0.0024, and 0.005 for H2SO4, NH3, HNO3, HCl, respectively). I checked the Sander et al. (2002) reference cited in the paper and found that the values used in this study are actually the lower limits reported for H2SO4, HNO3, and HCl condensing on highly concentrated sulfuric acid solution while the value for NH3 is the lower limit for it condensing on pure water. The authors need to explain the rationale and atmospheric relevance for these choices (assuming that accommodation coefficients for semi-volatile species are somehow needed in an equilibrium calculation, in the first place).

Reply:

The mass accommodation coefficient, by definition, indicates the mass fluxes of the condensable species from gas-phase to liquid/solid phases. However, some amounts of species may be evaporated back from liquid/solid phases to the gas-phase. Since by default the model treats the condensation of inorganic volatile gas species as irreversible process (no evaporation), the lower limit values of mass accommodation coefficients are used for these species to represent their net fluxes from the gas-phase to the liquid/solid phases. Such lower limit values correspond to uptake coefficients, which indicate the net fluxes and are smaller than mass accommodation coefficients.

To address the reviewer's comments, we have provided the rationale for the use of such lower limit values in the revised paper.

6. Please provide details on which new reactions and species are included in cloud (aqueous) phase chemistry. What numerical solver is used to integrate aqueous phase chemistry?

Reply:

The default aqueous-phase chemistry is based on Barth et al. (2000). Dissolution and dissociation of HNO_3 and HCl to produce NO_3^- and Cl^- in cloud water are added in the model based on Schwartz (1984), Marsh and McElroy (1985), and Seinfeld and Pandis (2006).

The concentration of H^+ (thus the pH value of the solution) is obtained by solving the electro-neutrality equation using the bisection method. The aqueous-phase chemistry of Barth et al. (2000) along with added aqueous dissolution and dissociation of HNO_3 and HCl is solved analytically.

This information has been added in the revised paper.

Reference cited:

Fountoukis, C and Nenes, A.: ISORROPIA II: a computationally efficient thermodynamic equilibrium model for K^+- Ca^{2+} -Mg^{2+} -$NH4^+$ -Na^+-SO_4^{2-}-NO_3^--Cl^--H_2O aerosols, Atmos. Chem. Phys., 7, 4639–4659, doi:10.5194/acp-7-4639-2007, 2007.

Fountoukis, C., Nenes1, A., Sullivan, A., Weber, R., Farmer, D., and Cohen, R. C., 2009, Thermodynamic characterization of Mexico City aerosol during MILAGRO 2006, Atmos. Chem. Phys., 9, 2141–2156, 2009.

Seinfeld, J. H. and S. N. Pandis, Atmospheric Chemistry and Physics: From Air Pollution to Climate Change. Hoboken, N. J, John Wiley, 1998.

Zaveri, R. A., R. C. Easter, J. D. Fast, and L. K. Peters, Model for Simulating Aerosol Interactions and Chemistry (MOSAIC), J. Geophys. Res., 113, D13204, doi:10.1029/2007JD008782, 2008.

Table 1. Comparison of ISORROPIA II box model results with those from ISORROPIA and AIM of Zaveri et al. (2008) under four conditions from Zaveri et al. (2008) [a]

	Case 1			Case 2			Case 8			Case 9		
	AIM[b]	ISO[b]	ISO II[b]	AIM	ISO	ISO II	AIM	ISO	ISO II	AIM	ISO	ISO II
NH_3	~ 220	275.7	275.7	~ 410	488.2	477.5	~ 420	549.8	505.8	~ 320	481.0	440.1
HNO_3	~ 340	348.2	348.2	~ 190	274.4	259.9	~ 140	236.0	164.1	~ 180	281.8	244.1
HCl	~ 4	4.1	4.1	~ 300	292.4	296.3	~ 310	330.9	358.9	~ 240	275.0	271.8
SO_4^{2-}					0.6	1.9	~ 26	12.0	78.9	~ 32	3.7	10.2
NH_4^+					3.3	18.1	~ 350	113.9	361.9	~ 250	16.0	56.8
NO_3^-					3.6	19.9	~ 240	76.5	148.3	~ 206	13.2	50.9
Cl^-					0.07	0.3	~ 68	13.5	55.8	~ 40	1.2	4.3
Na^+					1.5	5.8				~ 70	5.7	18.7
$(NH_4)_2SO_4$(s)	~ 120	118.4	118.4				~ 105	126.8	59.9	~ 6.8	1.3	1.3
NH_4Cl(s)					8.9	4.8	~ 105	146.3	76.0			
NH_4NO_3(s)	~ 50	28.8	28.8	~ 400	317.3	317.3		96.5	96.5		113.9	113.9
$NaNO_3$(s)				~ 17	18.0	16.3						
Na_2SO_4(s)				~ 40	40.2	39.0				~ 60	90.7	90.6
H_2O					3.7	15.3	~ 700	210.4	730.0	~ 600	24.1	83.5

[a] The unit is nmol m^{-3} for all values.

[b] ISO – ISORROPIA, ISO II – ISORROPIA II. AIM results are based on Zaveri et al. (2008). ISO results are based on ISORROPIA version 1.7 (3/15/06), which is very similar to ISORROPIA version 1.7 (03/26/07) used in Zaveri et al. (2008). ISO II results are based on the ISORROPIA II module used in our work.

Reply to Reviewer 4's Comments

This work describes several major model development and improvements in a recentlyreleased Earth system model, i.e., NCAR's CESM/CAM5.0. The paper focuses on model development effort and initial assessment of improved model treatments in representing the atmosphere. The model development includes a comprehensive gasphase chemical mechanism based on CB05 and detailed inorganic aerosol treatments that combine several nucleation parameterizations including advanced ion-mediated nucleation module of Yu (2010) and that simulate aerosol thermodynamics based on ISORROPIA II. A comprehensive model evaluation was performed for climatic/radiative variables and chemical concentrations and column mass abundances on global and several regional domains including US, Europe, and Asia using observations from global surface networks (including NCDC, GPCP, BSRN, and NOAA/CDC) and satellite datasets (including MODIS, CERES, TOMS/SBUV, MOPITT,GOME) as well as regional observational networks (including CASTNET, IMPROVE, STN over CONUS; the EMEP, BDQA, and AirBase over Europe; MEP of China, NIES of Japan, and TAQMN over East Asia). The simulation results were analyzed in detail and the improved model performance was linked clearly to specific model representations that were improved in this work, which helps the readers to better understand the benefits of such model improvements. It was found that CB05_GE with new and modified inorganic aerosol treatments in MAM7 is more accurate than simple gas chemistry coupled with default MAM7 and can predict many more gaseous species, and give improved performance for predictions of organic carbon and PM2.5 over CONUS, NH3 and SO42□ over Europe, SO2 and PM10 over East Asia, and cloud properties such as CF, CDNC, and SWCF. This work represents a significant contribution to the global air quality, climate, and Earth system communities and addresses several major model deficiencies and limitations in chemistry and aerosol treatments in current climate/Earth system models. It will thus potentially reduce the uncertainties of climate/Earth system predictions that are associated with those treatments in global climate and Earth system models. Although NCAR's CAM v5.0 represents the latest advancement in atmospheric model component in the contemporary Earth system model, it still uses simple gas-phase chemistry (only 8 chemical reactions among 6 gas-phase species) and highlysimplified thermodynamics that treats sulfate and ammonium only. In particular, it does not simulate nitrate and chloride which are important on a global scale. Modeling sulfate microphysics without considering these components introduces errors and uncertainties because it neglects microphysical interactions between components such as the interplay among sulfate, nitrate and ammonium (Yu et al., 2005) as well as sulfuric acid condensing onto sea-salt particles (O'Dowd et al., 1997). This work addresses the major model deficiencies in simulating chemistry and aerosols in CAM5.0 by incorporating the state of the science CB05_GE gas-phase chemical mechanism with 273 reactions among 93 gas-phase species and the latest version of ISORROPIA II. Incorporating CB05_GE enables an explicit simulation of many gas-phase species such as CO, O3, and HNO3 and also improves model performance for secondary PM species and PM precursors such as SO2 and NH3 in several regions. CB05_GE has been coupled with the most detailed aerosol module based on MAM7 in CAM5.0. To my best knowledge, CAM5.0 with the coupled CB05_GE/MAM7 represents the most advanced chemistry-aerosol treatments in current generation of Earth system models.

In a similar effort done by NCAR, MOZART version 4 (MOZART-4) (Emmons et al., 2010), an extended version of gas-phase chemistry used in Lamarque et al. (2012) has been incorporated into an offline version of CAM version 4.0 (older than CAM v5.0) driven by GEOS5 meteorological analyses. MOZART-4 was coupled to the simplified bulk aerosol module that cannot simulate aerosol size distribution. While MOZART_4E contains more explicit anthropogenic VOCs species than CB05_GE, it lumps monoterpenes as one species (i.e., _-pinene). By contrast, CB05_GE contains explicit treatments for more biogenic VOCs such as _-pinene, _-pinene, limonene, terpinene, and ocimine, thus enabling a more accurate treatment for secondary organic aerosol (SOA). It would be interesting to compare the two comprehensive gas-phase chemical mechanisms in the same CAM model to investigate their capabilities in simulating gas-phase chemistry and resulting secondary aerosol, in particular, SOA, on a global scale.

The incorporation of ISORROPIA II into CAM5.1 significantly advances aerosol treatments in current global and Earth system models. Although an older version of ISORROPIA was incorporated in several global models, most of which are offline-coupled meteorology-chemistry models such as GEOS-Chem (Bey et al., 2001) and the GISS Caltech (Liao et al., 2003); very few models are online-coupled models (e.g., the GUWRF/Chem (Zhang et al., 2012b)); and none of them are Earth system models that are much more complex than global or regional offline or online-coupled air quality models. Different from ISORROPIA that has been used in most regional air quality models such as CMAQ and CAMx and several global models, ISORROPIA II considers the effects of crustal species (e.g., magnesium, potassium, calcium) on inorganic aerosol thermodynamics that have been neglected in most regional air quality models and in nearly all global models. Not until recently, ISORROPIA II has been incorporated into CMAQ 5.0 (Appel et al., 2013). To my best knowledge, aerosol thermodynamics involving coarse particles has not been treated in nearly all regional and global models, not until recently, such effects were considered in CMAQ 4.7 and newer versions. Although this work only considers aerosol thermodynamics involving crustal species for fine-mode particles, it lays a foundation to further account for aerosol thermodynamics involving coarse particles and the effects of crustal species on coarse particles.

While climate models are typically run for 30-year or longer, very few work focusing on chemistry and aerosols can afford to simulate a 30-year period, due primarily to a large computational burden of long-term simulations using such air quality models. In this work, seven 1-year simulations were performed with various model treatments to illustrate the benefits of each improved model representation. I believe such 1-year model simulations are well sufficient for model development and initial assessments. It would be a natural extension for the authors to apply such a model with advanced chemistry and aerosol treatments for decadal simulations to demonstrate enhanced model capabilities in their future work.

In sum, this work represents a significant model development effort that deserves publication on ACP. The results are very interesting and promising. The paper is very well written and provides a very good documentation of their model development effort. The subject is appropriate to ACP. Therefore, I would strongly recommend its acceptance for publication on ACP after some minor modification. Several comments for improving the information content and presentation of the paper are listed below in specific comments.

Reply:

We thank the reviewer for positive comments.

Specific Comments
1. P27765-27767 for Tables 3-5: It will be better if the authors can add the model evaluation results for RMSE and NME for each variable.

Reply:

RMSE and NME were indeed calculated for all simulations but were not included in Tables 3-4 due to limited space. To address the reviewer's comments, we have added RMSE and NME in the Tables 6-7 from the three new 5-yr simulations.

2. P27777 for Figure 9, It will be better if the authors can compare the results for cloud and radiative variables for the summer time instead of whole year because the indirect aerosol forcing is more important during the summer.

Reply:

The results for cloud and radiative variables during Northern Hemisphere (N.H.) summer (JJA) are overall similar to those during the whole year. Although the magnitude of changes in CDNC, COT, and SWCF over specific regions is larger in N.H. summer-mean than annual-mean, the global mean changes in CDNC and SWCF are larger in annual mean than in N.H. summer-mean.

To address the reviewer's comment, we have added comparison for chemical, cloud and radiative variables for JJA mean in the revised paper, see Figures A2 and A3 in the supplementary material. We also evaluated the model performance during JJA and added this info in Tables A2 and A3 in the supplementary material.